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Surface Scaling Resistance of Concrete with Fly Ash From Co-Combustion of Coal and Biomass

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Abstract

The co-combustion of coal and biomass is connected with enforcing EU regulations concerning the application of renewable sources of energy. The more widely and efficiently that fuel is utilized the better this fact affects the ecology as well as the economy. The combustion of biomass and coal in thermal power stations gives a lot of benefits in comparison to coal burning only. The properties of fly ash from combustion of fuel mix including 80% of biomass and 20% of bituminous coal were described. Because of significant differences between fly ash tested and conventional fly ash, the ways of its application cannot be indicated without the risk of decrease in durability of concrete due to freezing and thawing. The results of frost resistance test of concrete with fly ash (*fab*) from co-combustion were presented. The frost resistance was evaluated based on the mass of scaled material related to the concrete surface subjected to 112 cycles of freezing and thawing in the presence of 3% NaCl solution. The statistical analysis of test results indicates that the introduction up to 25% of ground fly ash from co-combustion of biomass and coal to concrete, considering the air-entraining agent and proper curing conditions allows obtaining the durable concrete for structures subjected to external environmental influences

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Keywords: concrete, freeze-thaw resistance, fly ash, co-combustion of coal and biomass, air-entraining-agent, statistical analysis.

1. Introduction

Biomass, one of renewable energy sources, is an alternative to fossil fuels, which is used on an industrial scale. The EU regulations concerning the application of renewable sources of energy are the reason for their more significant contribution to the energy balance of many European countries [1]. The increased demands for energy have been forcing us towards more rational use of non-renewable sources of energy, together with the expansion of the renewable sources of energy, as well as the creation of ecologically and economically acceptable combination. According to Database Eurostat the average share of energy from renewable sources in gross energy consumption in the EU countries exceeded 14%. Considering the different geological as well as climatic conditions the renewable sources of energy varied from country to country – the energy from solid biomass predominates (from 34% in Italy to 99% in Estonia; in Poland – 95%). The application of biomass in power plants makes a significant contribution to the reduction of CO_2 emission [2], [3]. Among different technological solutions of energetic processing of biomass, the co-combustion of biomass and coal is of the most interest. The combustion of wood biomass and bituminous coal in existing large-scale boilers offers several advantages over great boilers fired exclusively with biomass, such as high electrical efficiency and lower investment costs. Substituting biomass for coal also reduces SO_2 emission through decreasing bound sulphur in the fuel [4].

The attempts of the multidirectional economical use of ashes from the combustion of different fuels have been made for years [5–9]. One of their main users is the industry of building materials. The way of the building materials manufacturing makes it possible to dispose a big amount of waste from other industrial sectors. Fly ash from hard coal is used extensively in concrete either as a separately batched material being an addition or as an ingredient in blended cement.

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It is used for economy, as it partially replaces an energy consuming material, the cement. Because of the wide availability and low cost, fly ashes are the most commonly used in the manufacture of cement-based materials to improve their microstructure [10–12]. Their main contribution, among others, is to the workability and the reduction of temperature rise in the fresh concrete as well as to the durability and long term strength development of hardened concrete. The beneficial effects of incorporating this material in concrete are widely discussed in the literature [5, 13, 14] but there is not enough available data concerning the properties of concrete with fly ash from co-combustion of coal and other fuels, e.g. biomass.

Practical application of new waste materials – the ash from co-combustion – demands the evaluation of their properties. The chemical composition of ash depends on the composition of coal, composition of biomass and its content in mixture of fuels as well as on the system of combustion [15]. Standard EN 450-1:2009 allows using the fly ash form co-combustion in concrete manufacturing, but it introduces the requirement for minimum content of coal in fuel mixture (80% by mass) and maximum content of ash from co-combusted materials (10%). Because of the effort to increase in biomass share in fuel mixtures, the most of fly ashes do not keep the requirements mentioned and the problem of further disposal of the growing stock of this kind of by-products still exists. The knowledge about them is usually insufficient among potential consumers.

The present study concerns the use of fly ash from co-combustion as a partial replacement of cement in concrete. However, that fly ash differs in physical and chemical properties from the traditionally used fly ashes. The useful properties of these materials have to be determined for every application. It is well known that the incorporation of fly ash changes the microstructure of cement composites, and that is why it influences their durability [16, 8, 17].

There are very few references concerning the frost resistance of concrete with fly ash from co-combustion of fuel mixture of high amount of biomass and the results presented are not conclusive. Johnson *et al.* [18] observed that the mortars containing 20% of fly ash from co-combustion of lignite and wood biomass were characterised by the frost resistance comparable with unmodified cement mortars. Wang *et al.* [8] pointed out that the concrete mixtures containing biomass fly ashes have higher demand of air entraining agents in order to achieve a specified volume of entrained air within the mix. Rajamma *et al.* [3] \and Safiuddin *et al.* [19] suggested to reduce the content of fly ash in concrete mix to 20% of cement mass to ensure the acceptable mechanical properties and durability. It is necessary to determine if this kind of ashes can be used in concrete elements subjected to external environmental influences.

The aim of the investigations was to evaluate the fly ash from co-combustion of hard coal and wood biomass (*fab*) influence on the surface scaling of concrete elements due to cyclic freezing and thawing in the presence of de-icing salt. Some studies on determining the content of fly ash and air-entraining agent, for obtaining concrete of required frost resistance, were presented in the paper.

2. Experimental investigation

2.1. Materials

The tests were carried on for concrete with Portland cement (CEM I 42,5 HSR NA), quartz sand, fraction 0÷2 mm, and the natural aggregate with maximum diameter of 8 mm. The aggregate mix consisted 40% of 0÷2 mm fraction, 25% of 2÷4 mm fraction and 35% of 4÷8 mm fraction.

The fly ash from co-combustion of bituminous coal (20%) and wood biomass (80%) in conventional boiler, in thermal power plant, was analyzed. The fly ash was used as a partial replacement of cement. The chemical composition of the cement and fly ash from co-combustion are presented in Table 1. The density of Portland cement was 3050 kg/m^3 and density of fly ash – 2684 kg/m^3.

Table 1. Chemical composition of Portland cement and fly ash tested

Component [% by	Portland cement	Fly ash
CaO	64.83	25.00
SiO_2	21.17	42.91
Al_2O_3	4.12	6.42
Fe_2O_3	5.08	2.80
MgO	0.80	2.45
SO_3	2.01	6.32
Na_2O_{eq}	0.53	4.40

The morphology of fly ash grains was determined using the environmental scanning electron microscope E-SEM (Nova Nano SEM 200) operated with a poor vacuum in specimen chamber. The specimens did not need to be coated with a layer of electrically conductive material. The observations were conducted with magnification from 500 to 10 000 times. The selected results of investigation were presented in Figs 1 and 2.

The fly ash from co-combustion contains small number of spherical grains. The plate and elongated shapes were also found but the most of grains had irregular or shapeless form. In chemical composition such components as carbon (in the form of loss on ignition), which can cause pozzolanic activity decrease, phosphor and significant amount of calcium have been observed.

Before introducing into concrete the fly ash was mechanically ground in a ball mill to the fine powder (< 0.063 mm). The grinding is the mechanical method of fly ash activation [4], [20].

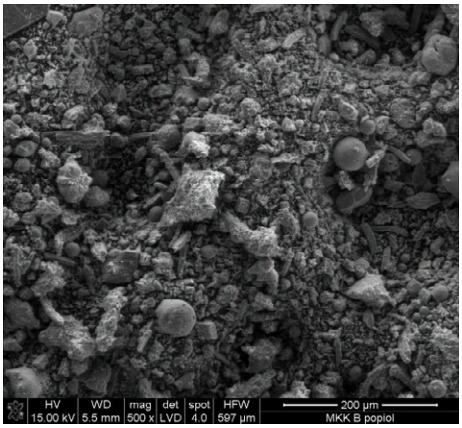

Fig. 1. E-SEM micrograph of fly ash from co-combustion; magnification 500×

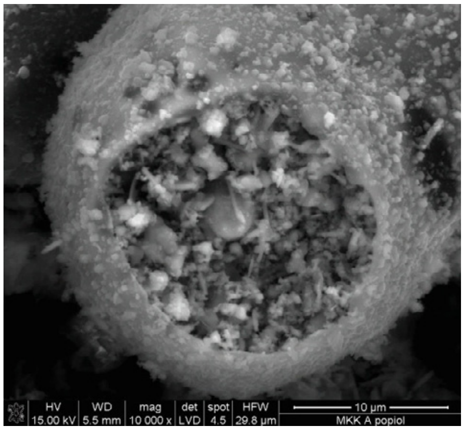

Fig. 2. E-SEM micrograph of fly ash from co-combustion; magnification 10 000×

Pozzolanic activity of ground fly ash was evaluated according to standard EN 450-1. The pozzolanic activity index was determined on the basis of the comparison of compressive strength (f_{cm}) of mortars containing 75% of cement and 25% of fly ash, and cement mortar without addition. The mortar mixture consisted of binder, water and fine aggregate in proportion 1:0.5:3. The value of pozzolanic activity index, determined after 28 and 90 days of curing, should exceed 75% and 85%, respectively. The values of pozzolanic activity after 28 and 90 days were 102% and 107%, respectively. Thus, the fly ash tested pointed out pozzolanic activity.

2.2. Mix proportions and specimens preparation

The concretes contained 0, 5, 15 or 25% of fly ash replacement related to cement mass (*fab/c*). The part of fly ash in mixture (40% for CEM I 42,5) was taken into account as binder and the remaining part – as filler, according to the standard EN 206-1:2003 [21].

Cement content in the control concrete was 350 kg/m^3. The water to binder ratio was equal to 0.50. The cement content and *w/b* ratio kept the requirements for concretes exposed to freezing and thawing with deicing salt solution according to EN 206-1.

The compositions of concrete mixes were given in Table 2.

Table 2. Compositions of the concrete mixes

fab/c	Cement [kg/m^3]	Fly ash [kg/m^3]	Water [kg/m^3]	Aggregate [kg/m^3]
0.0	350	–	175	1887
0.05	343	17.2	175	1873
0.15	330	49.5	175	1847

The air-entraining agent (AEA) content was 0.05 and 0.10% related to binder mass (100% of cement and 40% of fly ash). The concretes without AEA were also tested. The aim of introducing the AEA was to estimate the admixture efficiency in concrete mix containing fly ash considering the content of unburned coal [22]. The content of AEA was taking into account in total water content.

The test specimens were cast and compacted on a vibrating table in cubic moulds 100×100×100 mm. After 24 hours of curing, the specimens were removed from the mould and stored in tap water. The selected properties such as compressive strength, water absorbability and density were determined after 28 days of curing. The concretes were tested for compressive strength by the standard EN 12390-3:2002 [23].

The specimens for frost resistance testing were stored in water for 84 days, and for next 6 days – in the climatic chamber (20 ± 2 °C, 65% RH). The curing time in wet conditions was prolonged because of fly ash using [11], [24].

Every series was composed of 3 replicates. The 50 mm thick specimen was sawn from each cube perpendicular to the top surface, so that the saw cut for the freeze surface was located in the center of the cube.

The rubber sheet was glued to all surfaces of the specimen except the test surface. The edge of the rubber sheet reached 20 mm above the test surface. Then, all surfaces of the specimen except the test surface were thermally insulated. Top specimen surface was saturated with demineralized water during 72 hours. Directly, before the specimens were placed in the freezing chamber, the demineralized water was replaced with 3% NaCl solution. The freezing medium was prevented from evaporating by applying a flat polyethylene sheet. as shown in Fig. 3.

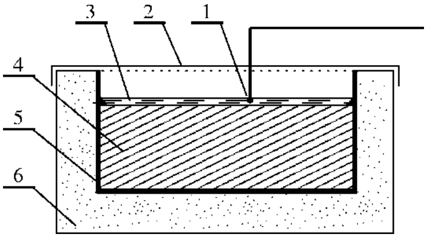

Fig. 3. The test specimen used for freeze-thaw test: 1 – temperature measuring device; 2 – polyethylene sheet; 3 – freezing medium; 4 – specimen; 5 – rubber sheet; 6 – thermal insulation

2.3. Test procedure

The concrete resistance to scaling due to cyclic freezing and thawing with de-icing salt saturation (3% NaCl solution) was determined using the procedure described in CEN/TS 12390-9:2007 (slab test) [25]. It is the most sever method of frost resistance test [26], [27]. The behavior of concrete under field exposure conditions is simulated, the type of deterioration is found in real structures and the test results evaluation is quantitative.

In the freezing chamber with temperature and time controlled cooling and heating system, the specimens were subjected to repeated freezing and thawing. During the test cycle, the temperature in freezing medium changed within the graph shown in Fig. 4.

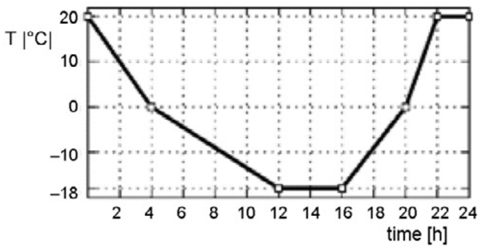

Fig. 4. The time-temperature cycle in the freezing medium

Every 7 days NaCl solution was exchanged. The material that had scaled from the test surface was collected and dried to constant weight. The amount of the scaled material per unit area after n cycles m_n was calculated for each measuring occasion and each specimen.

The conformity criteria for concretes according to SS 13 72 44 (1995) [28] – Borås method were presented in Table 3.

Table 3. Criteria for scaling resistance evaluation

Scaling resistance	Requirements
Very good	$m_{56} < 0.10$ kg/m^2
Good	$m_{56} < 0.20$ kg/m^2 or $m_{56} < 0.50$ kg/m^2 and $m_{56}/m_{28} < 2$ or $m_{112} < 0.50$ kg/m^2
Acceptable	$m_{56} < 1.00$ kg/m^2 and $m_{56}/m_{28} < 2$ or $m_{112} < 1.00$ kg/m^2
Inacceptable	$m_{56} > 1.00$ kg/m^2 and $m_{56}/m_{28} > 2$ or $m_{112} > 1.00$ kg/m^2

The mean mass of scaled material after 28, 56 and 112 cycles was used for evaluating the scaling resistance qualitatively.

3. Results and discussion

3.1. Selected properties of concrete with fly ash from co-combustion

The test results of the selected properties of concretes were summarized in Table 4.

Table 4. Selected properties of concretes with fly ash addition after 28 days of curing: compressive strength f_{cm}, water absorbability n_w, density ρ

AEA content [% c.m.]	*fab/c* –	f_{cm} [MPa]	n_w [%]	ρ [kg/m^3]
0.0	0.0	59.7	3.50	2242
	0.05	52.2	3.55	2252
	0.15	49.1	3.20	2298
	0.25	45.2	3.33	2329
0.05	0.0	57.0	3.42	2255
	0.05	51.0	3.40	2240
	0.15	48.6	3.29	2272
	0.25	42.5	3.15	2300
0.10	0.0	55.2	3.28	2265
	0.05	49.4	3.17	2237

The addition had no significant influence on material's density and water absorbability.

The fly ash addition, in considered range of *fab/c* values, caused the decrease in compressive strength determined after 28 days of curing. The rate of strength development in concretes with fly ash was slower in comparison to that of control specimens, particularly at early ages. The changes in concrete, caused by chemical reactions in system cement-fly ash-water, appear usually after prolonged time, and their rate depends on pozzolanic activity of ash as well as on curing conditions [2], [5].

3.2. Analysis of scaling resistance results

The variations in the mass of scaled material with fly ash content related to cement mass, air entraining agent content as well as number of cycles for concretes tested were presented in Fig. 5.

The scaling resistance was evaluated qualitatively considering the criteria given in Table 3. The resistance of concretes without air-entraining agent (Fig. 5a) was hardly acceptable according to criteria from Table 3. The value of m_{56} was greater than 0.50 kg/m^2 for all concretes tested. The rate of scaling accumulation was comparable independently of the fly ash content in concrete, but the content of addition had an influence on cumulative mass of scaled material.

The introduction of 0.05% of air-entraining agent related to binder mass, caused the improvement in frost resistance of concrete without fly ash as well as the resistance of concretes containing up to 15% of mineral addition (Fig. 5b). The rate of growth in scaling was the greatest for concrete of *fab/c* = 0.25 and the resistance of this concrete was acceptable only.

The increase in AEA content up to 0.10% related to binder mass (Fig. 5c) made it possible to achieve very good resistance to scaling for concretes tested, except for the concrete containing 25% of fly ash, for which the average mass of scaling, after 56 cycles was equal to 0.15 kg/m^2. After 112 cycles, the mass of scaled material was 0.284 kg/m^2 for concrete of *fab/c*=0.25 and 0.09 ÷ 0.166 kg/m^2 for the rest of concretes.

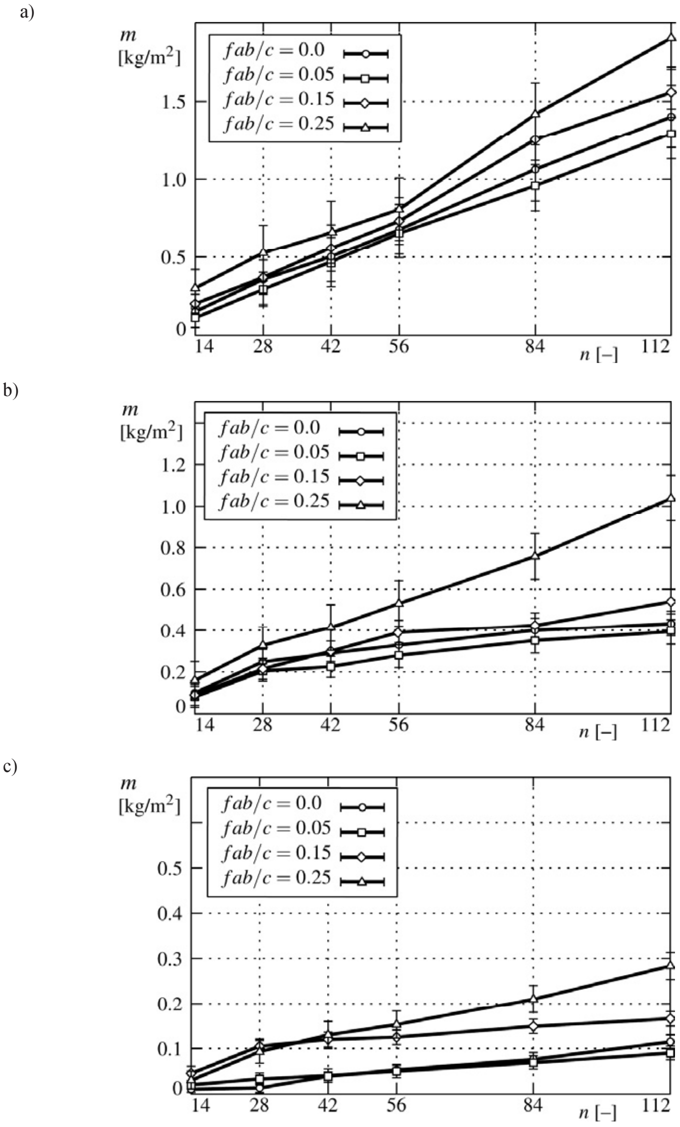

Fig. 5. Mean mass of scaling residue *m* vs. *fab/c* value as well as number of cycles *n* for different content of AEA: a) concretes without AEA; b) concretes with 0.05% AEA; c) concretes with 0,10% AEA

Regarding the complex influence of factors considered on surface scaling resistance of concrete the statistical analysis of test results was performed [29], [30]. The mathematical model describing the process of accumulation of scaled material in dependence on selected factors was developed.

The effect of the air-entraining agent content (factor X_1), fly ash content (factor X_2) and the number of freeze/thaw cycles (factor X_2) on the mass of scaling (variable Y) was analyzed.

The range of variation and the levels of the factors considered were as follows:

– factor X_1: $i_1 = 0.0$ (–1); $i_2 = 0.05\%$ (0); $i_3 = 0.10\%$ (1),
– factor X_2: $fab/c_1 = 0.0$ (–1); $fab/c_2 = 0.05$ (–0.333); $fab/c_3 = 0.15$ (0.333); $fab/c_4 = 0.25$ (1),
– factor X_3: $n_1 = 28$ (–1); $n_2 = 56$ (–0.333); $n_3 = 84$ (0.333); $n_4 = 112$ (1).

The normalized values of factors were given in parentheses. The normalization of the independent variables made it possible to evaluate the quantitative effects of selected factors on dependent variable (mass of scaling) analyzing the values of regression coefficients determined for independent variables.

The function $Y = f(X_1, X_2, X_3)$ was used to describe the factor space. The values of equation's coefficients were determined using the least-squares method.

The design of experiment was worked out allowing obtaining the adequate mathematical description of regression function. The design for three variables composed of N=48 trials was used. Every series was composed of three replicates. Carrying on the experiment the rule of randomization of tests sequence was kept, taking into account the specificity of test method.

Preliminary analysis of test results (Fig. 5) showed the scatter of Y values in each trial.

Before the main analysis the Cochran's G test was used as a homogeneity test. The results of scaling residue measurements for concretes after 28, 56, 84 and 112 freeze-thaw cycles were taken into account. The test showed that for 48 means and the degrees of freedom $d_f = 2$, the calculated value equals:

$$G_{ijk\,\max} = S^2_{ijk\,\max} / \Sigma S^2_{ijk} = 0.1278, \tag{1}$$

for level of significance $\alpha = 0.05$, is less than $G_{0.05,2,48} = 0.1398$ [29], thus the compared variances are homogeneous and the test results are reproducible.

On the basis of the input normalized data the mathematical model describing the accumulation of scaling (function $\hat{Y}$) was developed:

$$\hat{Y} = 3.65 - 4.23X_1 + 1.25X_2 + 2.71X_3 - 0.47X_1X_2 - 2.70X_1X_3 + 0.82X_2X_3 + 1.04X_1^2 + 1.56X_2^2. \tag{2}$$

The response surface function was characterized by coefficient of determination $R^2 = 0.970$. The significance of the equation variables was verified using t-Student test according to Brandt [29].

The basic effects and the factors' interactions were analyzed. From practical point of view, the fly ash content is the most important factor. Thus, the relationship obtained (2) was presented graphically in the form of response surface and isolines for concretes without air-entraining agent as well as for concretes containing 0.05% of AEA in dependence on *fab/c* value (Fig. 6a and 6b). For concretes with 0.10% of AEA, the influence of fly ash content was slender.

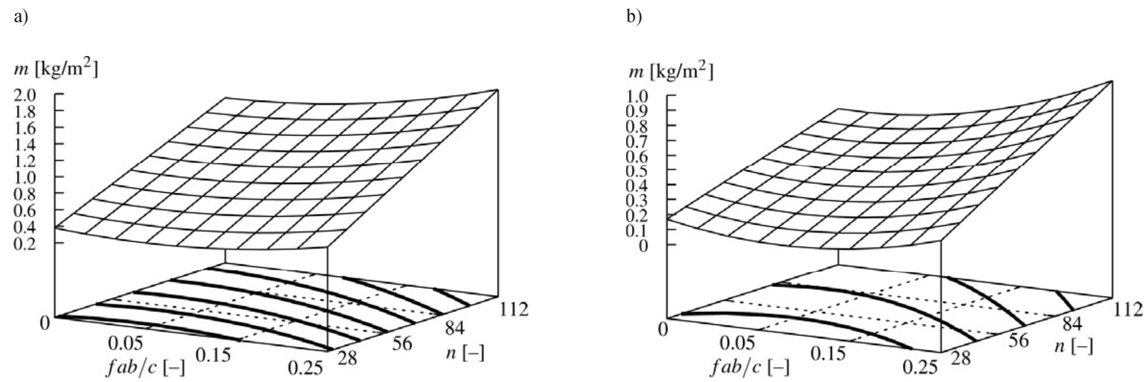

Fig. 6. Graphical presentation of the response surface (2): a) concretes without AEA; b) concretes with 0.05% AEA

In the considerations presented the positive effect means the effect causing the increase in mass of scaling, and the negative effect means the decrease in mass of scaled material.

The analysis of the model (2) showed the main influence of air-entraining agent content (factor X_1) on mass of scaling. The linear negative as well as little square positive effect of this factor were identified. The presence of AEA caused the limitation of scaling, but its influence was unequal. The introducing of 0.05% of AEA caused 59% diminution of mass of scaling in comparison to the concrete without AEA. The increase in AEA content up to 0.10% of binder mass, caused the scaling limitation about 87% in comparison to the concrete containing 0.05% of air-entraining agent.

Significant negative effect of simultaneous action of X_1X_3 confirmed that the increase in AEA content (X_1) makes the influence of cyclic freezing and thawing (X_3) less effective. Little negative effect of X_1X_2 testifies that the influence of fly ash content (X_2) on mass of scaling decreases when the content of AEA increases.

The positive linear effect of cyclic freezing and thawing was found. The synergetic effect of X_2X_3 means that the simultaneous action of these factors influences the accumulation of scaling stronger than everyone separately.

The increase in the fly ash content caused the increase in mass of scaled material. The influence of fly ash content was confirmed by positive linear and positive square effect according to regression model.

4. Conclusions

The pozzolana activity of fly ash has mainly been pointed out as well as the possibility to use this addition as a concrete component. The fly ash addition, in considered range of *fab/c* values, has no significant effect on density and water absorbability of concrete but the mineral addition reduces the compressive strength determined after 28 days of curing.

The aim of this study was to assess the durability of fly ash from co-combustion concrete subjected to cyclic freezing and thawing in the presence of deicing salt solution.

Elaborated and verified statistical model was used for detailed assessment of the complex influence of fly ash content and air-entraining agent content on scaling resistance of concrete. The analysis of the relationship obtained showed that fly ash has an effect on deterioration of concrete surface, although its content was limited to 25% of cement mass. However, the proper air entrainment improves the resistance to scaling and makes it possible to achieve good resistance for all mixes tested.

The regression equation makes the estimation of concrete scaling resistance possible and this way, allows choosing the optimal composition of concrete for specific applications considering the life cycle of structure. The knowledge of basic properties of fly ash from co-combustion of coal and biomass is fundamental to its effective usage in building industry.

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